

Waveform controls on few-cycle pulses enable measurement of attosecond dynamics.

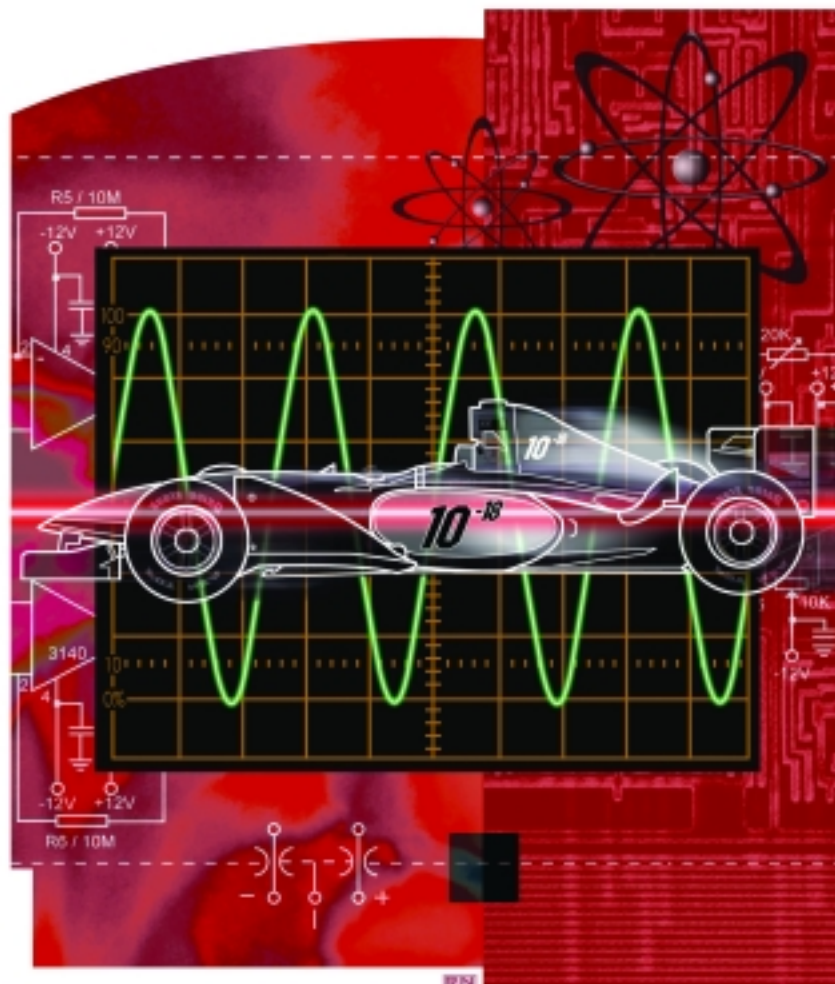


ILLUSTRATION BY RANDALL NELSON

PULSED LASERS *Speed Up*

The electric field oscillations of visible light change their sign about 10^{15} times per second and, therefore, the field strength changes from zero to maximum in less than a femtosecond. By precisely controlling these oscillations within a very short laser pulse, we have demonstrated an apparatus capable of measuring atomic processes with an accuracy of approximately 100 as ($1 \text{ as} = 10^{-18} \text{ s}$).

The process begins with an intense, few-cycle (about 5-fs) laser pulse with a controlled waveform that creates a 250-as soft-x-ray pulse by ionizing atoms. This sub-femtosecond burst subsequently excites the electronic system of atoms. The electric field oscillations of the same 5-fs laser pulse are then used to probe the time structure of electron emission accompanying the impulsive x-ray excitation. This new measuring technique is capable of tracing hyperfast dynamics in the electron shells of atoms on the time scale of the Bohr

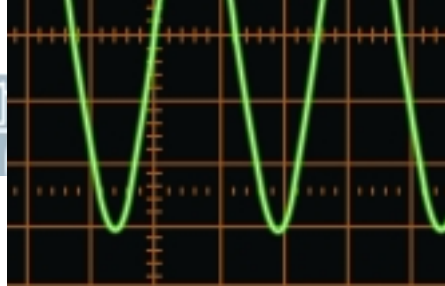
By Matthias Uiberacker, Vienna University of Technology, and Ferenc Krausz, Max-Planck-Institute for Quantum Optics and Vienna University of Technology

orbit time in hydrogen (approximately 150 as) for the first time. We can also use this method to measure the duration of sub-femtosecond laser pulses.

Electrons in Motion

With state-of-the-art microscopes, scientists can observe atoms at rest. If, however, the atoms are in motion, very short light pulses are needed to reconstruct the motion from a series of snapshots. While an exposure time of less than 0.001 s can produce a sharp image of a tennis ball in flight, capturing fast atomic events requires light pulses of just a few femtoseconds in duration. To monitor the electrons themselves, scientists would have to shorten the light pulse by another hundred to thousand times.

Electrons are important objects of study because they control much about a material's properties, including its reaction to other chemicals. As stated above, electrons change from one energy state to another within tens to thousands of



attoseconds. In the process, these electrons can cause atoms originally bound to a molecule to fly apart or emit UV radiation or x-rays. These processes are of fundamental significance for controlling chemical reactions and synthesizing new materials. Until recently, however, pulses on the order of a few femtoseconds were the shortest illumination sources available for the study of dynamic events.

Time-resolved measurement of attosecond transients calls for new tools and measuring techniques. The process begins with initiating the microscopic process under study. This is typically accomplished by imparting energy to the atom to set its interior in motion and must take place within a fraction of the duration of the entire process under study.

In the case of electron imaging, energy input has to occur within a fraction of a femtosecond. For this purpose, we have developed a laser-driven, coherent, soft-x-ray source that emits individual bursts with a duration of 250 as. The sub-femtosecond x-ray flash excites the electron shell of the irradiated atoms. The implications of this excitation include the emission of primary (photo) and secondary (Auger) electrons from the atoms. The duration and evolution of this electron emission provide direct information on the evolution of the excitation and relaxation processes in the electron shell.

Light-Field-Controlled Streak Imaging

We have now developed a method for measuring these processes in a time-resolved fashion. This involved reverting to a well-known concept, streak imaging.^{1,2} Until recently this method was used exclusively for measuring the duration of short light flashes. The light flash impinging on the streak imager's metal plate knocks some electrons loose. These electrons are then accelerated by a static electric field towards a fluorescent screen. Before they hit the screen, they are deflected with another field that increases linearly with time. The temporally varying deflection "streaks" the point of impact of the electrons on the screen. The spatial width of this streak image (Δx) is directly proportional to the duration of the electron emission, i.e. the duration of the light flash (Δt). The faster the deflecting field varies, the shorter are the pulses that can be recorded. State-of-the-art streak cameras attain resolutions approaching 100 fs.

The novelty of the new method lies in the fact that a light field, which varies millions of times faster than a standard electric field, deflects the electrons. For this purpose, we supplement the attosecond x-ray flash irradiating the atoms

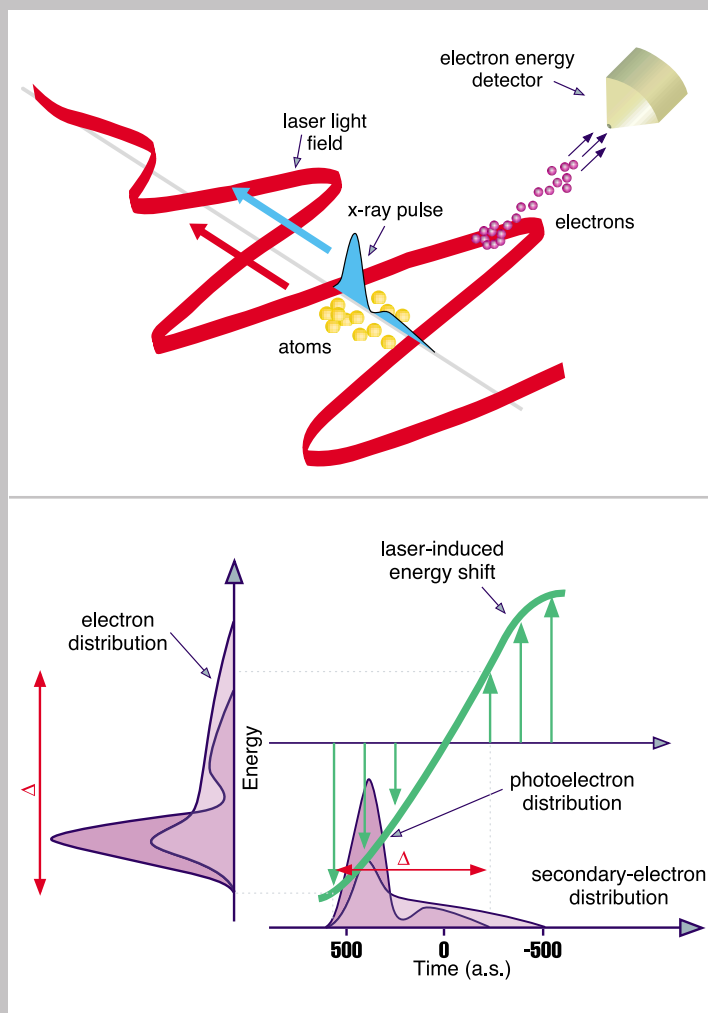


Figure 1 In the light-field-controlled streak camera (top), atoms are exposed simultaneously to a sub-femtosecond x-ray flash (blue curves) and a laser pulse consisting of a few well-controlled field oscillation periods (red curves). Depending on the time of birth of an electron within the half period of the light field oscillations, the kinetic energy of the electron increases or decreases during the few femtoseconds required for the laser pulse to pass the atoms. In a projection of the measured energy to hypothetical temporal emission profile of photo and Auger electrons (bottom), the width (ΔE) and shape of the electron energy distributions mirror the duration (Δt) and evolution of the emission within a sub-femtosecond time window.



When Ultrafast is Not Fast Enough

If you are fishing for a physicist, the best bait can be an intriguing problem. Tibor Juhász, then a young researcher at the Institute for Experimental Physics of the Budapest University of Technology (Budapest, Hungary), posed such a problem to a potential doctorate student, Ferenc Krausz, in the mid-1980s. Juhász asked Krausz to develop a technique to measure the pulse duration of a single shot from a picosecond flashlamp-pumped neodymium-doped glass laser. Several years later, Krausz would co-invent the chirped multilayer mirror to control pulse dispersion in femtosecond lasers, a major contributor toward the acceptance of ultrafast lasers in laboratories around the world and the resultant understanding of dynamic molecular events.



Krausz completed his master's degree in electrical engineering in Budapest before earning a doctorate in quantum electronics at the Vienna University of Technology (VUT; Vienna, Austria), where he subsequently taught. He is about to leave VUT upon becoming a director at the Max-Planck-Institute for Quantum Optics (Garching, Germany) and a chair of experimental physics at the University of Munich (Munich, Germany). Juhász went on to pioneer laser eye surgery with femtosecond pulses at IntraLase (Irvine, CA).

Krausz has not stopped with the chirped mirror. In this edition of *oemagazine*, Krausz discusses how his team, working with researchers at the University of Bielefeld (Bielefeld, Germany), has managed to lock, measure, and reproduce light pulses with sub-femtosecond precision, opening up the world of attosecond science and enabling some of the first direction measurements of electrons in motion within their atomic shell.

In a previous life, Krausz enjoyed reading, music, and sports. Now, though? "If I wish to spend some time with my kids [and in the lab], the time left for hobbies quickly approaches zero," he laughs.

Zero. Now that's fast. —Winn Hardin



with an intense laser pulse comprising just a few oscillations (see figure 1). The x-ray flash excites the atoms, which then emit electrons. The electrons ejected in the direction of the electric field polarization of the laser light pulse are detected. Depending on the time of their emission within half the oscillation period of the laser light, the electrons undergo a change of velocity; in the case illustrated, the electrons emitted first are decelerated, while those released on termination of the x-ray flash are accelerated.

This method allows us to separately detect the successively emitted electrons, not spatially on a screen this time but alongside one another on the energy scale. The width ΔE and shape of the measured energy distribution of the electrons reflect the duration and evolution of the electron emission, in the same way that the spatial distribution represents this information in conventional streak imaging. In this case, however, deflection occurs within half a light period, which opens the way to measurement in the attosecond regime.

In order to implement this concept of a light-field-controlled streak camera successfully in practice, we have to accomplish three tasks: generate a very short x-ray flash, precisely control the oscillations of the deflecting light pulse,

and synchronize the exciting x-ray flash to the oscillations of the deflecting pulse with attosecond precision. Accomplishing this task requires intense laser pulses consisting of a few, well-controlled field oscillation cycles.

Controlled Waveforms

A laser system producing such waveform-controlled, few-cycle pulses was demonstrated recently by our group in collaboration with Theodor Hänsch and colleagues from the Max-Planck-Institute for Quantum Optics (Garching, Germany).³ In this system, a titanium-doped sapphire (Ti:sapphire) oscillator delivers pulses with precise control of the phase of the electric field oscillations with respect to the peak of the pulse envelope—the so-called carrier-envelope (C-E) phase. Pulses having the same C-E phase, and hence the same waveform, are selected from the 80-MHz train at a 1-kHz repetition rate and amplified in a Ti:sapphire multipass, chirped-pulse amplifier system to a pulse energy of about 1 mJ. Phase-locked loops ensure C-E phase stability at the output of the oscillator and after amplification. Finally, a hollow-fiber/chirped-mirror compressor stage reduces the pulse duration down to 5 fs at a carrier wavelength of approximately 750 nm (corre-

sponding to an oscillation period about 2.5 fs).

The waveform-controlled 5-fs, 0.3-mJ pulses are subsequently sent into an evacuated beamline (see figure 2). We focus the laser beam into a neon-gas jet, inside of which the laser's electric field induces gigantic atomic dipole oscillations by pulling an electron out of the atom as far as several nanometers and smashing it back towards its core half a cycle later. The oscillations are not sinusoidal but contain very-high-frequency components that extend into the extreme-UV and soft-x-ray spectral regions.

In a laser field containing many oscillation cycles, the oscillations are repeated quasi-periodically, resulting in the emission of a series of high-energy bursts of sub-femtosecond duration as well as high-order harmonics of the laser radiation in the spectral domain.⁴⁻⁶ A few-cycle laser field induces only a few dipole oscillations, each with a different amplitude. The oscillation with the highest amplitude has been predicted to produce a single burst in the spectral range of the highest emitted photon energies.⁷ First evidence for the emission of an isolated sub-femtosecond pulse from this interaction was reported in 2001.⁸ The reproducible generation of single sub-femtosecond pulses was not

Figure 2 In the system, 5-fs visible laser pulses focused into a neon gas jet create a few high photon energy x-ray bursts; plasma clouds effuse from the interaction region (see inset). The excitation and x-ray pulses pass through a high-pass filter (zirconium foil), which blocks both laser light and low-energy x-ray photons. Subsequently, the multilayer mirror unit selects only the highest-energy spectral components of the x-ray radiation, which are confined to a single burst. The mirror unit can also introduce a delay between x-ray burst and laser pulse, and focus the two beams into a second gas target for implementing the light-field-controlled streak camera measurements.

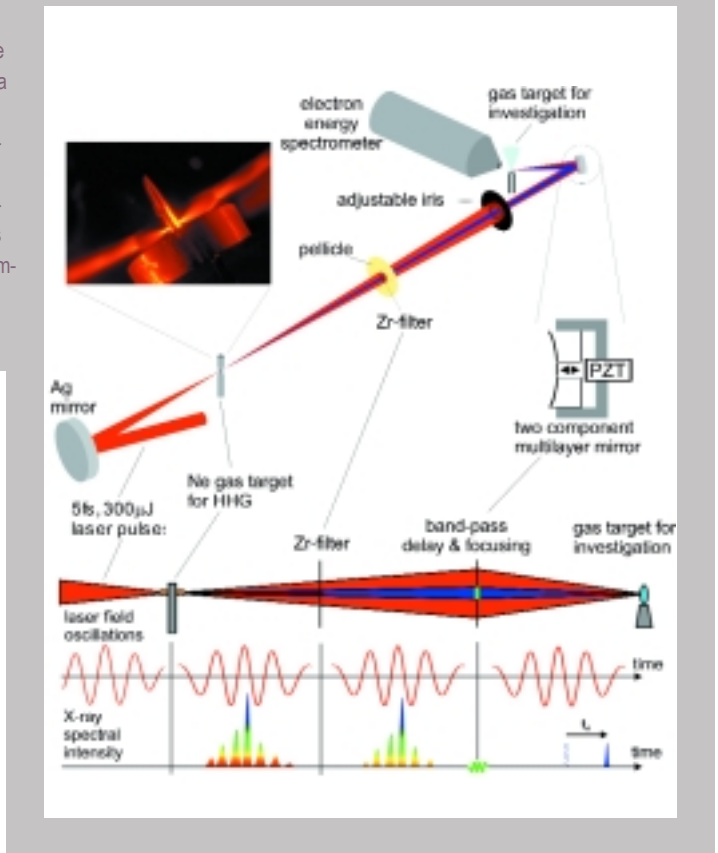
possible until 2003, when researchers created a method to precisely control the giant atomic dipole oscillations with intense waveform-controlled, few-cycle laser pulses.

The X-ray Source

The ensemble of these tiny antennas emits x-rays collinearly with the laser beam. Thanks to reduced beam divergence, we can spatially separate the soft x-rays from the laser beam using a thin metal foil inserted into the beam to block the laser light in the region of the soft-x-ray emission. Subsequently, the two beams pass through an iris for controlling the energy of the laser pulse and hit a spherical molybdenum/silicon (Mo/Si) multilayer mirror that reflects the laser beam and acts as a band-pass filter for radiation at around 93 eV (13 nm) with a bandwidth of 9 eV.

The focusing mirror consists of two parts: a 3-mm-diameter inner part and an outer part with a 3-mm hole. The inner part is mounted on a piezo-controlled translation stage with nanometer precision to ensure spatial overlap of the two beams and introduce a delay between the laser and the x-ray pulse with attosecond precision. The Mo/Si multilayer mirror only reflects the highest-energy x-ray photons. Based on our few-cycle excitation conditions, we predict these x-ray photons to be temporally confined to a single burst.

Although the intensity of the 5-fs pulse leads to x-ray emission, the use of waveform-controlled light excites the atomic oscillations in a manner that is precisely reproducible from one laser shot to the next. This consistency results in x-ray bursts with reproducible parameters, including duration, energy, and timing relative to the laser field. In this way, the system appears to fulfill the conditions stated above and we can implement measurements with the light-field-controlled streak camera.



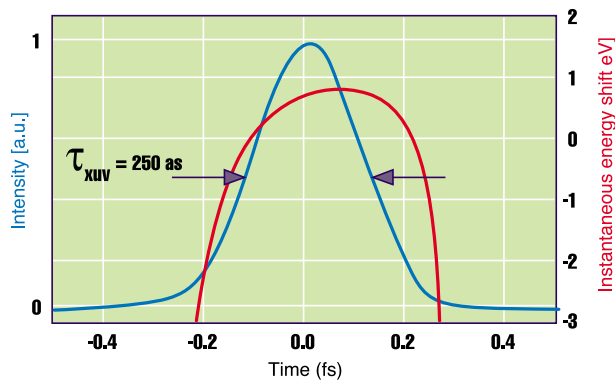


Figure 3 Plot of temporal intensity profile (blue) and energy sweep (red) of a single x-ray burst evaluated by light-field-controlled streak camera shows attosecond pulse.

To achieve these streak measurements, the Mo/Si mirror focuses the few-cycle laser pulse along with the x-ray flash into a second ensemble of atoms, which are impulsively excited by the x-ray burst. The electrons ejected from the atoms upon this excitation are deflected—or, in our case, accelerated or decelerated—by the few-cycle laser field. We measure the energy of the electrons with a time-of-flight (TOF) spectrometer.

The evolution of the photoelectron emission ejected by the x-ray flash reflects the time structure of the exciting x-ray flash, while the evolution of an Auger electron provides information on the relaxation processes that occur in the electron shell in the wake of the flash-like excitation. By measuring the emission time of the photoelectrons using the streak method, we have been able to determine both the duration and the chirp of the sub-femtosecond x-ray pulse (see figure 3). The measurements also yielded conclusive evidence regarding the isolated nature of the burst by confirming the absence of satellite bursts with notable fractional energy.

When applied to secondary electrons, our apparatus will allow direct measurement of the evolution of processes inside the electron shell of atoms with a resolution of approximately 100 as, which is within the Bohr orbit time.⁹ For the first time, researchers have a chance of presenting

experimental answers to questions related to possible ways of producing an effective x-ray laser, as well as fathoming the processes that govern the creation and destruction of chemical bindings, including the creation of novel new materials. **oe**

Matthias Uiberacker is a graduate student at the Photonics-Institute of the Vienna University of Technology, Vienna, Austria;



and Ferenc Krausz is the director of the Max-Planck-Institute for Quantum Optics, Garching, Germany, and a professor at the Vienna University of Technology. For questions, contact Krausz at +49 89 32905 602, +49 89 32905 314 (fax), or ferenc.krausz@mpq.mpg.de.

References

1. D. Bradley, B. Liddy, and W. Sleat, *Opt. Commun.* 2, p. 391 (1971).
2. M. Schelev, M. Richardson, and A. Alcock, *Appl. Phys. Lett.* 18, p. 354 (1971).
3. A. Baltuska, et al., *Nature* 421, p. 611 (2003).
4. P. Paul, et al., *Science* 292, p. 1689 (2001).
5. Y. Mairesse, et al., "Attosecond Synchronisation of High-Harmonic Soft X-Rays," *Science* 302, to be published.
6. P. Tzallas, et al., "Direct observation of attosecond light bunching," *Nature* 425, in press.
7. T. Brabec, F. Krausz, *Review of Modern Physics* 72, p. 2 (2000).
8. M. Hentschel, et al., *Nature* 414, p. 509 (2001).
9. M. Drescher, et al., *Nature* 419, p. 803 (2002).